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## Hybrid process of coagulation/flocculation with *Moringa oleifera* followed by ultrafiltration to remove *Microcystis* sp. cells from water supply

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### Abstract

In Brazil and other countries, the large biomass of monospecific crops and the need to enhance plant growth by the extensive use of fertilizers have caused a fast eutrophication of rivers and reservoirs that has resulted in a high growth in the population of cyanobacteria, of which several species have been described as producers of toxins capable of causing death of domestic and wild animals, in addition to problems to human health. The objective of the present work was then to evaluate the efficiency of *Moringa oleifera* seeds as coagulant in removing cells of the cyanobacterium *Microcystis* sp., apparent color, and turbidity from water in association with ultrafiltration. For the experiments, deionized water was artificially contaminated with *Microcystis* sp. cells from a culture. Mixing was carried out until samples with initial turbidity of 350 and 450 NTU were obtained to simulate high turbidity water with bloom of cyanobacteria. After being prepared, the samples were subjected to the processes of (1) coagulation/flocculation with moringa seeds (CFM), (2) UF, and (3) combined coagulation/flocculation with moringa solution followed by ultrafiltration (CFM-UF). The CFM process was carried out with a solution of 1% moringa seeds at concentrations of 25 to 300 mg/L. Mixing was promoted in a jar test device. Poly(ether sulfone) membrane with retention of 50 kDa and operating pressure of 2 bar was used for the UF process. In the CFM process, turbidity removal ranged from 49 to 97.4%, color removal varied between 39 and 99.2%, and removal of *Microcystis* sp. cells ranged from 20 to 91%. Applying the analysis of variance to the results, optimal moringa concentrations of 175 and 250 mg/L were obtained for water samples of 350 and 450 NTU respectively. These concentrations were used in the combined CFM-UF process. *Microcystis* sp. cells were not detected in treated water after UF and CFM-UF processes. Color and turbidity removal was above 99% for both processes. For the permeate flux, the combined CFM-UF process and the UF process alone presented average relative fluxes of 40% and 29.4%, respectively. Membrane fouling was 78.5% for UF and 70% for CFM-UF. This way, it can be said that coagulation/flocculation with moringa can be applied to high turbidity water, with high removal of color, turbidity, and *Microcystis* sp. cells, reaching more

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than 99% when this process was combined with ultrafiltration. In addition, the combined process showed higher permeate flux and slightly lower fouling percentage when compared with the UF process alone.

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**Keywords:** *Microcystis* sp.; *Moringa oleifera*; ultrafiltration; water treatment

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## 1. Introduction

Contamination of water resources, especially in areas with inadequate sanitation and water supply, has become a risk factor for health problems, with water playing a role as a vehicle for transmission of biological agents (viruses, bacteria, and parasites).

In Brazil and other countries, the large biomass of monospecific crops and the need to enhance plant growth by the extensive use of fertilizers have caused a fast eutrophication of rivers and reservoirs that has resulted in a high growth in the population of cyanobacteria, of which several species have been described as producers of toxins capable of causing death of domestic and wild animals, in addition to problems to human health [1, 2].

In addition to these microorganisms, many other impurities can harm human health if not reduced or eliminated. However, these impurities do not approach each other, it is necessary to add a coagulant. Chemical coagulants are the most used and among them the most common is aluminum sulfate, since it is cheap and easily obtained. Recently, much attention has been drawn on the extensive use of alum. According to Driscoll and Letterman [3], the utilization of alum has raised a public health concern because of the large amount of sludge produced during the treatment and the high level of aluminum that remains in the treated water. McLachlan [4] discovered that the intake of a large quantity of alum salt may cause Alzheimer disease.

One alternative that arises in this context is the use of natural coagulants that have advantages over chemical coagulants since they are biodegradable and non-toxic, and produce sludge in less quantity and with lower metal content. Seeds of *Moringa oleifera* Lam (moringa), which contain active agents with coagulant properties, is an example of natural coagulant.

The use of coagulants for drinking water treatment, in spite of being efficient in the removal of most contaminants, is not able to generate water of high potability standards, which leads to the necessity of the simultaneous use of other techniques. Membrane filtration technique is already widely recognized and can be implemented in combination with coagulation processes. Today, membrane filtration technology is recognized by the water industry as a very attractive process for producing drinking water. Membranes are physical barriers that are able to efficiently remove suspended particles and colloids, turbidity, bacteria, algae, parasites, and viruses for clarification and disinfection purposes, as well as to control trihalomethane precursors. In comparison with conventional processes such as coagulation, flocculation, sedimentation and/or flotation, and rapid or slow sand filtration, membrane filtration technology has many advantages such as superior quality of treated water, much greater compactness, easier control of operation and maintenance, use of fewer chemicals, and lower production of sludge. Communities are increasingly looking to membrane filtration as a safer treatment alternative.

To overcome the problems caused by natural organic matter (NOM), for example fouling and permeate flux decrease, in membrane filtrations applications, conjunctive use of coagulation and membranes is becoming more attractive for water treatment because the coagulation is an opportunity to join NOM with other particles present in water before NOM reaches the membrane surface. The NOM found in the liquid leads to membrane fouling, flux reduction and inferior effluent quality. Therefore, the application of coagulants for the raw water pretreatment may bring about an improvement in permeates quality. This is

very important, especially in the case of drinking water production.

Membrane processes are now economically attractive for large facilities using good quality surface water. Currently, the objectives are to extend membrane technology processes to poorer quality water for the removal of color, taste, dissolved organic matter, and disinfection by-products.

Considering this context, the objective of the present work was then to evaluate the efficiency of moringa seeds as coagulant in removing cells of the cyanobacterium *Microcystis* sp., apparent color, and turbidity from water in association with ultrafiltration.

## 2. Materials and methods

### 2.1 Water samples

For the experiments, deionized water was artificially contaminated with *Microcystis* sp. cells from a culture. Mixing was carried out until samples with initial turbidity of 350 and 450 NTU were obtained to simulate high turbidity water with bloom of cyanobacteria.

After being prepared, the samples were subjected to the processes of (1) coagulation/flocculation with moringa seeds (CFM), (2) UF, and (3) combined coagulation/flocculation with moringa solution followed by ultrafiltration (CFM-UF).

### 2.2 Coagulation/flocculation process with moringa (CFM)

Moringa coagulant solution was prepared and used the same day. Mature moringa seeds from the Federal University of Sergipe (UFS) were used as raw material. The seeds were manually removed from the dry pods and peeled. To prepare the 1% stock solution of moringa (concentration of 10,000 mg/L), 1 g of peeled seeds was crushed and added to 100 mL of distilled water. Subsequently, the solution was stirred for 30 min and vacuum filtered [5]. From the 1% stock solution, moringa solutions were prepared with different concentrations: 25, 50, 75, 100, 125, 150, 175, 200, 225, 250, 275, and 300 mg/L. Mixing was promoted in a jar test device under the following conditions: coagulation time of 100 s; rapid mixing gradient of  $85 \text{ s}^{-1}$ ; flocculation time of 600 s, and slow mixing gradient of  $40 \text{ s}^{-1}$  [5].

The optimal concentration of moringa coagulant solution – with the highest removal of *Microcystis* sp., turbidity, and apparent color – was determined for each water sample and then used to perform the combined CFM-UF process.

### 2.3 Ultrafiltration (UF) and combined coagulation/flocculation with moringa solution followed by ultrafiltration (CFM-UF).

To membrane filtration process, poly(ether sulfone) membrane with molar retention of 50KDa was tested using the principle of crossflow filtration, with an operating pressure of 2 bar. The final produced water quality was assessed considering the efficiency of removal of apparent color, turbidity and cyanobacterium cells.

Filtration tests were performed to characterize the flow of pure water through the membranes, using deionized water. Flux was calculated using Equation 1, where  $f_{\text{permeate}}$  is the permeate flux,  $m$  is the mass of collected water,  $\rho_{25^\circ\text{C}}$  is water density at  $25^\circ\text{C}$ ,  $\Delta t$  is the time interval during which water was collected, and  $A_m$  is the filtering area of the membrane.

$$F_{\text{permeate}} = m / \rho_{25^\circ\text{C}} \cdot \Delta t \cdot A_m \quad (1)$$

The removal efficiency for each parameter analyzed in the different treatment processes was calculated from Equation 2, where  $C_i$  and  $C_f$  are the initial and final concentrations, respectively, for each parameter.

$$\% \text{Removal efficiency} = (C_i - C_f / C_i) \cdot 100 \quad (2)$$

Deionized water (DW) fluxes were determined before each experiment ( $J_i$ ) and after the filtration of solutions SW, CFQ and CFS ( $J_f$ ) to determine the fouling of the membrane. The percentage of fouling (%F) was calculated according to Equation 3, proposed by Balakrishnan et al. [6], using the steady-state flux values, which assume that the flux tends to constant values. The percentage of fouling represents a decrease in the deionized water flux after tests with contaminated water.

$$\%F = [(J_i - J_f) / J_i] \cdot 100 \quad (3)$$

In Equation 3, %F is the percentage of membrane fouling,  $J_i$  is the initial water flux obtained in the first filtration with deionized water and  $J_f$  is the final water flux obtained by filtration of deionized water after the filtration of surface water.

## 2.4 Analytical methods

Apparent color and turbidity were measured in a HACH DR/2010 spectrophotometer, according to the procedure recommended by Standard Methods [7], pH was measured by a Digimed DM-2 pH meter according to the manufacturer's methodology.

Removal degree of *Microcystis* sp. cells was monitored by the Utermöhl method, according to methodology described by Lund et al. [8], which involves the counting of sediment organisms in a special chamber using an inverted microscope.

## 3. Results and discussion

### 3.1 Results of CFM process

Results of apparent color, turbidity and *Microcystis* sp. cells removal and pH values of treated water with coagulation / flocculation process with moringa (CFM) are shown in Table 1.

Table 1. The efficiency of *Microcystis* sp. cells, apparent color and turbidity removal and pH values of treated water with different concentrations of moringa seeds solution.

Initial turbidity (NTU)	Removal efficiency (%)	Moringa concentration in the water samples (mg/L)											
		25	50	75	100	125	150	175	200	225	250	275	300
350	Turbidity	49.4	62.8	70.8	75.9	55	66.5	93.7	95	96	95.8	96.4	92.5
	Apparent color	49.0	54.8	62.3	69.8	75.5	86.3	99.2	94.0	95.4	95.3	95.6	91.3
	<i>Microcystis</i> sp.	60.9	74.2	72.7	26.8	20.7	35.4	69.2	79	33.3	46.5	72.7	47.7
	pH	7.78	7.87	8.15	7.77	7.81	7.74	7.82	7.75	7.73	7.76	7.73	8.13
450	Turbidity	53.0	61.0	75.0	79.9	92.0	94.0	97.2	97.4	97.2	97	96.7	94.7
	Apparent color	39.0	47.8	61.6	68.5	88.3	91.5	96.1	96.1	96.4	96.0	95.6	92.8
	<i>Microcystis</i> sp.	63.3	61.2	49.9	51.7	87.4	82.5	84.9	83.2	83.5	91.0	88.9	89.6
	pH	8.21	7.77	7.65	7.61	7.66	7.63	7.59	7.62	7.66	7.61	7.56	7.64

In table 1, to CFM process, turbidity removal ranged from 49 to 97.4%, color removal varied between 39 and 99.2%, and removal of *Microcystis* sp. cells ranged from 20 to 91%. Applying the analysis of variance to the results, optimal moringa concentrations of 175 and 250 mg/L were obtained for water samples of 350 and 450 NTU respectively. These concentrations were used in the combined CFM-UF process.

Good results have been observed for the removal of cyanobacteria by coagulation/flocculation, depending on the characteristics of the organic matter present in water, the cyanobacteria species, and the type and concentration of coagulant [9]. The use of natural coagulants such as *Moringa oleifera* Lam (moringa) replacing chemical coagulants have been a viable alternative for cyanobacteria removal, due to the use of a small dosage of biopolymers, which can greatly reduce the consumption of chemical coagulants, with the advantage of generating less sludge, being biodegradable, not changing the pH of the water, and providing good removal of color and turbidity.

### 3.2 Results of UF and CFM-UF process

In Table 2 are presented the removal efficiencies of parameters apparent color, turbidity, *Microcystis* sp. cells and pH values of treated water by UF and CFM-UF processes.

Table 2. Efficiency of turbidity, apparent color and *Microcystis* sp. cells removal and pH values of treated water by UF and CFM-UF processes.

Initial turbidity (NTU)	Removal efficiency (%)	Water treatment process	
		UF	CFM/UF
350	Turbidity	99.89	99.62
	Apparent color	100.0	99.58
	<i>Microcystis</i> sp.	ND*	ND
	pH	7.33	8.42
450	Turbidity	99.77	99.80
	Apparent color	99.79	100.0
	<i>Microcystis</i> sp.	ND	ND
	pH	7.48	8.00

\*ND: not detected

*Microcystis* sp. cells were not detected in treated water after UF and CFM-UF processes, so it can be assumed that the concentration of cells was below the detection limit, in agreement with other studies that used UF for removing cyanobacteria cells [10, 11]. Color and turbidity removal was above 99% for both processes. It can be seen that the removal efficiency for the parameters analyzed are very similar and for the water sample 450 NTU turbidity, it is observed that the combined process showed slightly better results when compared to UF process without pretreatment.

For the permeate flux, the combined CFM-UF process and the UF process alone presented average relative fluxes of 40% and 29.4%, respectively (Fig. 1).

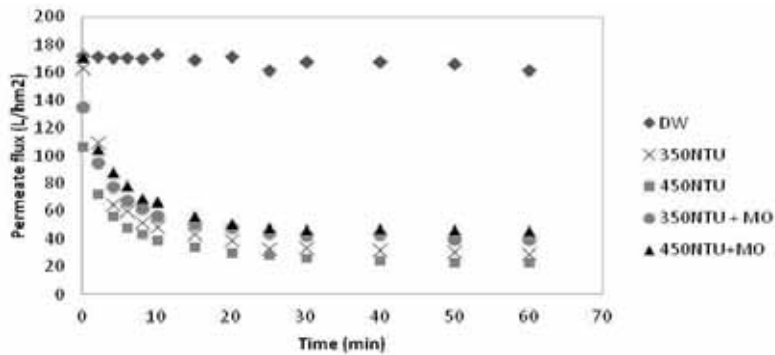


Fig. 1. Permeate flux with deionized water (DW) and raw water with initial turbidity 350 and 450 NTU in the UF (a) and CFM-UF (b) processes

Membrane fouling was 78.5% for UF and 70% for CFM-UF (Fig. 2) and the predominant fouling mechanism was cake filtration according to the model of Duclos-Orsello et al. [12] (Fig. 3).

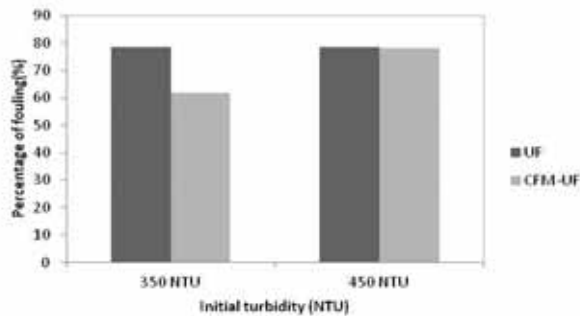


Fig. 2. Percentage of fouling for the UF process with raw water and water coagulated/flocculated with moringa (CFM) followed by UF with initial turbidity of 350 and 450 NTU

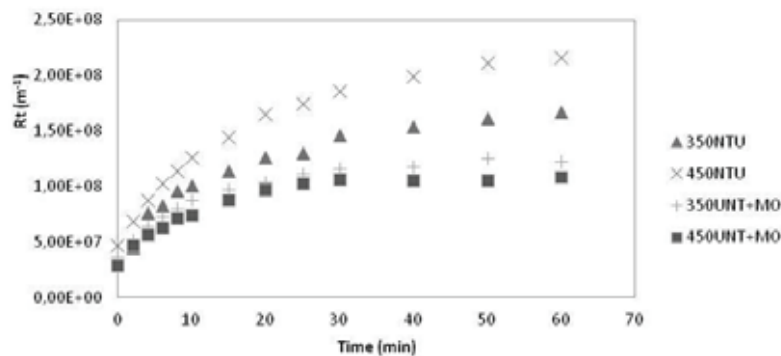


Fig. 3. Total Resistances for the UF process with raw water and water coagulated/flocculated with moringa (CFM) followed by UF with initial turbidity of 350 and 450 NTU

For UF membrane, the curve remains concave downwards for all experiments, due to lower porosity of

the UF membrane, it can be said that the permeation of particles into the membrane pores does not occur, the particles being deposited on the surface of membrane over time, characterized by the mechanism of fouling by cake formation [12].

Thus, it can be said that combined processes showed lower %F and flux slightly greater when compared to membrane filtration processes isolated. This improvement of MF and UF performance in combined processes are observed in other studies [11,13].

#### 4. Conclusions

This way, it can be said that coagulation/flocculation with moringa can be applied to high turbidity water, with high removal of color, turbidity, and *Microcystis* sp. cells, reaching more than 99% when this process was combined with ultrafiltration. In addition, the combined process showed higher permeate flux and slightly lower fouling percentage when compared with the UF process alone.

Therefore, the use of moringa seeds can be considered a promising step to improve the process of coagulation/flocculation of water associated with membrane filtration, seeking, among other things, the removal of *Microcystis* sp. cells. It is a promising technique for improving the quality of water intended for human consumption.

Bulleted lists may be included and should look like this

- Coagulation/flocculation with moringa can be applied to high turbidity water, with high removal of color, turbidity, and *Microcystis* sp. cells
- The combined process showed higher permeate flux and slightly lower fouling percentage when compared with the UF process alone.
- Therefore, the use of moringa seeds can be considered a promising step to improve the process of coagulation/flocculation of water associated with membrane filtration, seeking, among other things, the removal of *Microcystis* sp. cells

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